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Revision 3

Beryllium Waste Transuranic Inventory in the Subsurface Disposal Area, Operable Unit 7-13/14

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August 2005

Idaho Cleanup Project

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Assistant Secretary for Environmental Management
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ABSTRACT

This report documents new characterization data applied to inventory estimates associated with irradiated beryllium disposed of in the Subsurface Disposal Area, a radioactive waste landfill in the Radioactive Waste Management Complex at the Idaho National Laboratory. Between 1970 and 1993, irradiated beryllium from the Advanced Test Reactor, Engineering Test Reactor, and the Materials Test Reactor was buried in the Subsurface Disposal Area. Radionuclide inventories associated with these beryllium blocks were based on earlier characterization methods and measured data.

Chemical assay data obtained from irradiated beryllium now stored in the canal at the Advanced Test Reactor were used to validate the calculated inventories associated with beryllium waste buried in the Subsurface Disposal Area. Computational models used in this study and the data on which they were based are described in this report. Also discussed is irradiated beryllium not yet disposed of that will eventually require disposal. Additional research into shipment data has resulted in some changes from the previous issue of this report to both the reported radionuclide inventory at time of disposal and the disposal locations of the irradiated beryllium.

These changes in radionuclide inventory and disposal locations of the irradiated beryllium supercede information in previous versions of this report.

EXECUTIVE SUMMARY

Beryllium is used as a neutron reflector in some research reactors. At the Idaho National Laboratory's Reactor Technology Complex (formerly Test Reactor Area), beryllium has been used for core components in three reactors: the Advanced Test Reactor (ATR), the Engineering Test Reactor (ETR), and the Materials Test Reactor (MTR). Between 1970 and 1993, beryllium irradiated in those three facilities was buried as radioactive low-level waste in the Subsurface Disposal Area (SDA) at the Radioactive Waste Management Complex, part of the Idaho National Laboratory. New information about radionuclide concentrations in irradiated beryllium from beryllium now being stored in the canal at ATR indicates that previous characterization of irradiated beryllium is incorrect. These new data have reduced uncertainty in beryllium characterization data compared to earlier data.

Though a number of radioisotopes were modeled and documented in this study, the analysis focused on C-14 and several transuranic (TRU) waste isotopes identified as potential risk drivers for Operable Unit 7-13/14. The following conclusions derived from this study:

- C-14 inventory in the buried MTR, ETR, and ATR beryllium was approximately 92.5 Ci at time of disposal.
- This study verifies that TRU concentrations are present in beryllium from ATR (previously not known), and that these concentrations exceed 100 nCi/g for TRU waste.
- Approximately 2.47 Ci of TRU isotopes are attributable to disposal of beryllium from MTR, ETR, and ATR. Estimated TRU inventories in buried beryllium are small compared to total TRU inventories in the SDA.
- Computer modeling for characterization of beryllium reflector blocks and outer shim control cylinders (OSCCs) from ATR was validated by comparing model concentrations to measured concentrations for several isotopes at two sample point locations on Block 010R. The data presented in this report represent the best-estimate characterization now available for approximately 40 radionuclides considered in this analysis.
- This study validates the computer models and the basis for segmented and single-block computer models to determine accurately the total block inventory of TRU isotopes and other radionuclides of interest.
- Based on calculation by the segmented three-dimensional model, more than 96% of the beryllium block mass exceeds 100 nCi/g. In addition, nearly 50% of the block mass exceeds 400 nCi/g.
- The single-block model calculated a more conservative TRU total block average concentration than the segmented model and associated detailed analysis.
- TRU concentration is a dynamic quantity and can be accurately determined from computer analysis. Primary variables that influence TRU inventory are irradiation time, initial uranium inventory, neutron flux, and decay time. Following irradiation, TRU isotopes significantly increase in concentration because of beta decay of non-TRU isotope Pu-241 into TRU isotope Am-241.
- The segmented model provides insight into the three-dimensional distribution of important isotopes within a typical beryllium block from ATR, and also is a powerful tool for determining alternative or representative sampling locations for future sampling.

- Elemental assay data presented in this report represent known, best-estimate information about chemical impurities in beryllium reflector material used at the INL Site and in the industry.

- This investigation indicates that two of the Core 2 blocks are still in the ATR canal and that six Core 2 blocks were disposed of on or about October 14, 1982 in the SDA. This study has confirmed that nine OSCCs came from Cores 1 and 2, but which core positions these OSCCs occupied are still unknown.

Recommendations

Although the Materials Test Reactor and Engineering Test Reactor characterization data in this report apply specifically to beryllium disposed of from these reactors, with slight revision the data could be applied to beryllium reflector materials and other core components remaining in the Engineering Test Reactor and Materials Test Reactor to support future waste disposal for facility deactivation, decontamination, and decommissioning. Reports by Kaiser et al. (1982) and Rolfe and Wills (1984) should be revised to reflect information in this report.

All beryllium reflector material now stored in the Advanced Test Reactor canal or generated in the future should be modeled to estimate the total radiological inventory to support future waste path planning. In addition, modeling should be expanded to estimate the radiological inventory that will result from continued operation of the Advanced Test Reactor Critical facility.

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ACRONYMS

ATR	Advanced Test Reactor
ATRC	Advanced Test Reactor Critical (facility)
ETR	Engineering Test Reactor
ETRC	Engineering Test Reactor Critical (facility)
INL	Idaho National Laboratory
KBI	Kawecki Berylco Industries
LLW	low-level waste
MCNP4B	Monte Carlo N-Particle Version 4B
MFC	Material and Fuels Complex
MOCUP	MCNPB4-ORIGEN2 Coupled Utility Program
MTR	Materials Test Reactor
ORIGEN2	Oak Ridge Isotope GENeration and Depletion Code Version 2
OSCC	outer shim control cylinder
OU	operable unit
RI/FS	remedial investigation/feasibility study
RWMC	Radioactive Waste Management Complex
RWMIS	Radioactive Waste Management Information System
SDA	Subsurface Disposal Area
TRU	transuranic

Beryllium Waste Transuranic Inventory in the Subsurface Disposal Area, Operable Unit 7-13/14

1. INTRODUCTION

Beryllium is used as a neutron reflector in some research reactors. At the Idaho National Laboratory Site (INL; formerly the Idaho National Engineering and Environmental Laboratory), beryllium has been used in three reactors: the Advanced Test Reactor (ATR), the Engineering Test Reactor (ETR), and the Materials Test Reactor (MTR) located in the Reactor Technology Complex (formerly the Test Reactor Area). Between 1970 and 1993, beryllium irradiated in those three facilities was buried as radioactive low-level waste (LLW) in the Subsurface Disposal Area (SDA) of the Radioactive Waste Management Complex (RWMC) at the INL Site. New data about radionuclide concentrations in irradiated beryllium now being stored in the ATR canal indicate that previous characterizations of irradiated beryllium were incorrect. These new data have reduced the uncertainty in the beryllium characterization data compared to earlier characterization data.

1.1 Purpose

The purpose of this report is to provide estimates of radionuclide inventories associated with irradiated beryllium disposed of in the SDA. The inventory will be used to support the development of the comprehensive remedial investigation/feasibility study (RI/FS) for Waste Area Group 7[a], Operable Unit (OU) 7-13/14. The RI/FS will ultimately provide the basis for remedial decision-making under the Federal Facility Agreement and Consent Order (DOE-ID 1991).

1.2 Scope

New beryllium characterization methods and resulting data have been developed to support planning for disposal of beryllium reflector waste stored in the ATR canal. These methods and data have also been used to revise and improve the accuracy of the radiological source term for irradiated beryllium waste buried in the SDA that came from the ATR, ETR, and MTR reactors. The new information has been incorporated into this report to provide improved data for characterizing the radiological properties of both irradiated beryllium waste that has previously been disposed of and possible future disposals. The computational models central to this characterization, their inputs and outputs, and the measurements on which they were based are discussed in some detail. Also included is a discussion of the irradiated beryllium not yet disposed of that will eventually require disposal. This material is in the ATR canal; in the ATR, MTR, and ETR reactors; and waiting or planned for placement in the ATR.

This revision of the report addresses modifications in the radionuclide inventory of the irradiated beryllium at time of disposal. The original model calculations have not been changed; however, the radiological inventory at time of disposal has been modified because of locating additional shipment data identifying changes in the disposal dates and the disposal locations of the irradiated beryllium since the previous version of this report was published. The radiological inventory presented in this revision

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a. The Federal Facility Agreement and Consent Order lists 10 waste area groups for INL. Each waste area group is subdivided into OUs. The RWMC is identified as Waste Area Group 7 and originally contained 14 OUs. Operable Unit 7-13 (transuranic pits and trenches RI/FS) and OU 7-14 (Waste Area Group 7 comprehensive RI/FS) were ultimately combined into the OU 7-13/14 comprehensive RI/FS for Waste Area Group 7.

supersedes the data in the previously published report. In addition, beryllium grouting activities that have been undertaken since the report was published previously are also briefly discussed.

1.3 Background

The data in this report support the OU 7-13/14 comprehensive RI/FS required under the Federal Facility Agreement and Consent Order (DOE-ID 1991) and the Comprehensive Environmental Response, Compensation, and Liability Act (42 USC § 9601 et seq. 1980). The INL Site, RWMC, and other facilities are shown in Figure 1-1. A map of RWMC showing the SDA is shown in Figure 1-2.

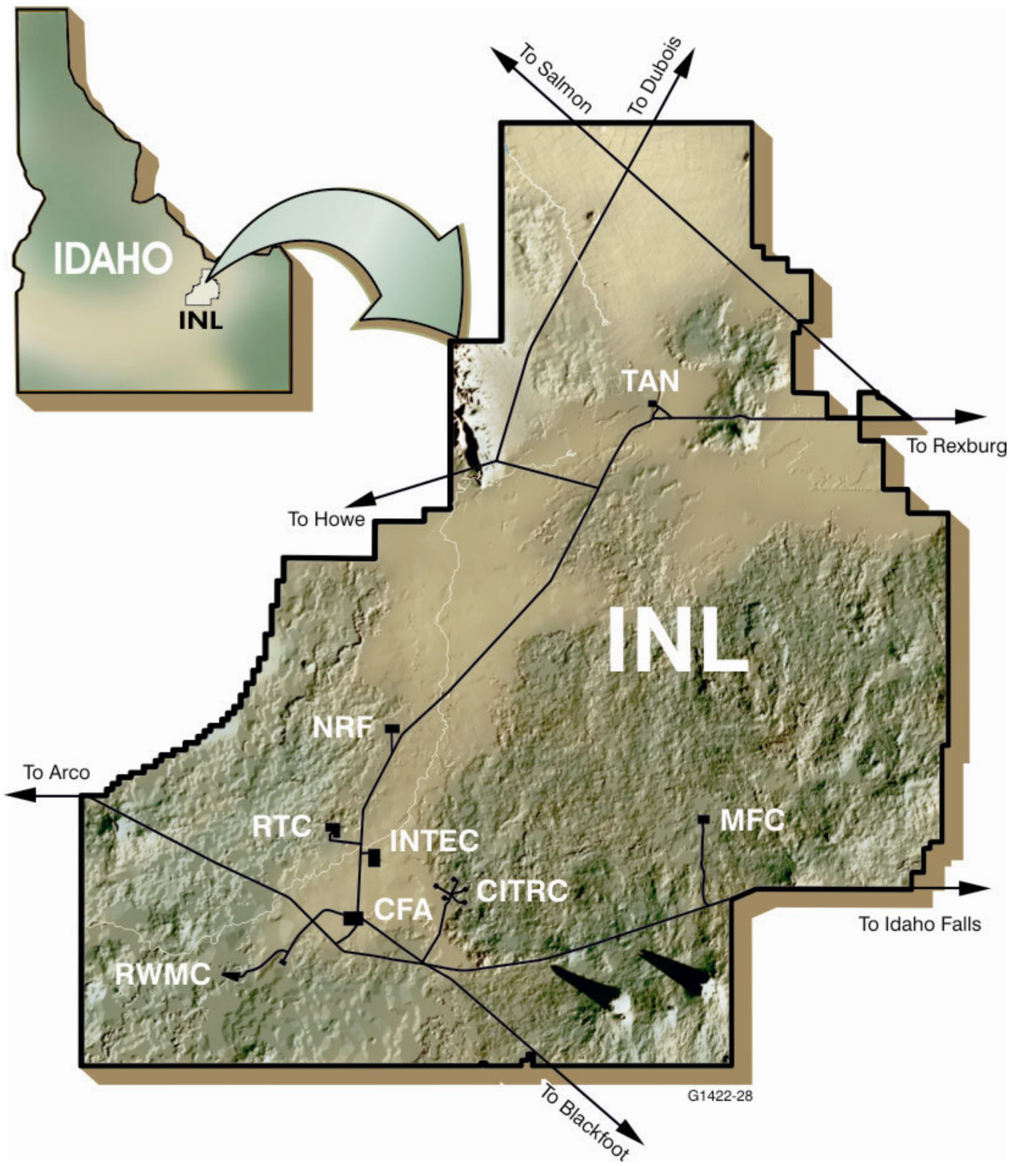

Figure 1-1. Relief map of the Idaho National Laboratory showing the location of the Radioactive Waste Management Complex and other facilities.

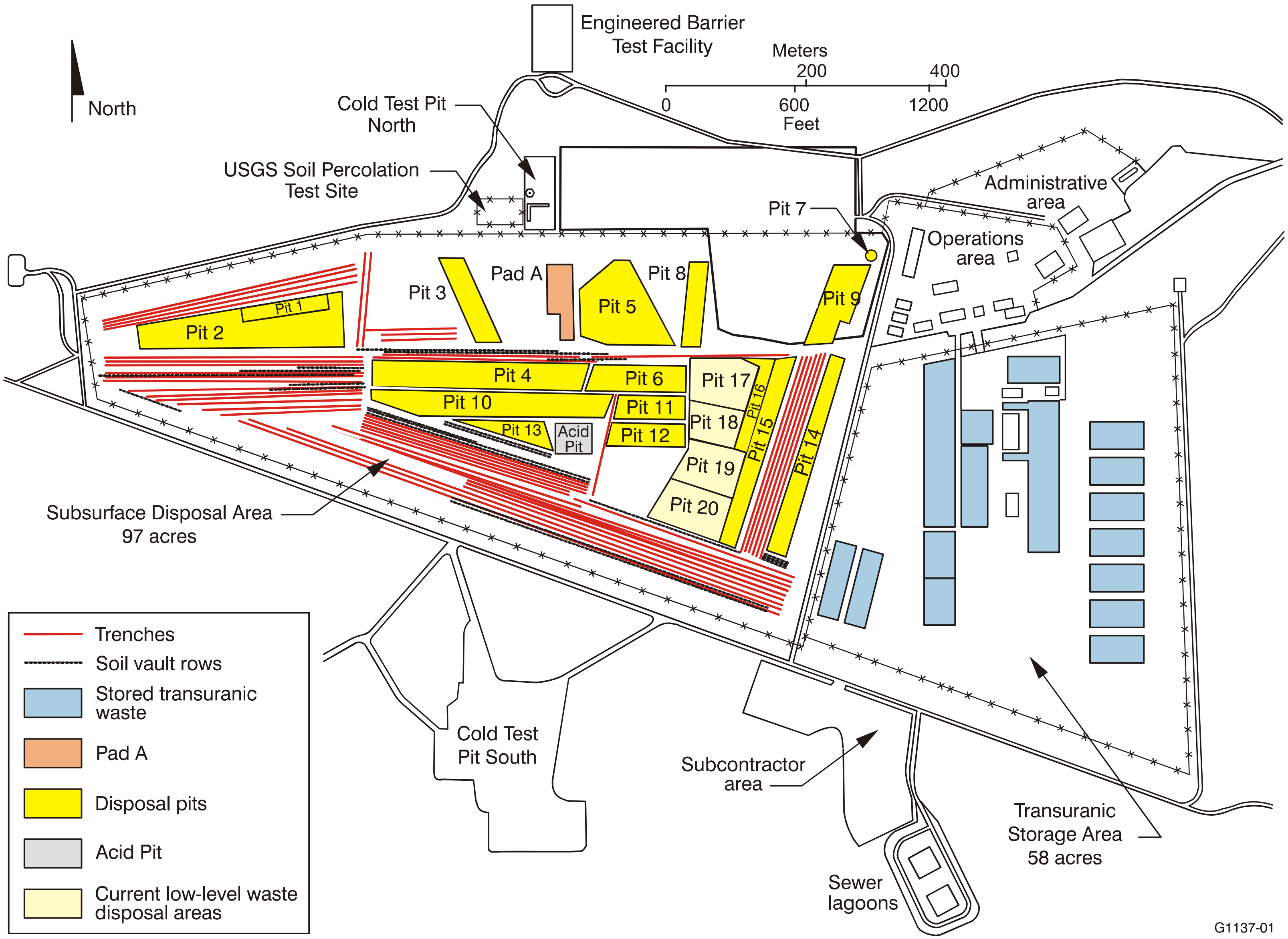

Figure 1-2. Map of the Radioactive Waste Management Complex showing location of the Subsurface Disposal Area.

The boundary of Waste Area Group 7 is defined as the RWMC fence. The SDA is a fenced portion within RWMC that includes numerous pits, trenches, and soil vaults where transuranic (TRU) waste and LLW have been buried since 1952. Disposal of hazardous material in the SDA ceased in 1983. Since 1983, only LLW meeting the requirements of INL waste acceptance criteria (DOE-ID 2002a) has been disposed of in the SDA active LLW disposal pit.

The MTR, ETR, and currently operating ATR reactors at INL Site use beryllium reflectors. The ATR reflector consists of a set of eight beryllium blocks and 16 outer shim control cylinders (OSCCs). In the ETR, the reflector is essentially four blocks of beryllium that surround the core, while in the MTR, the reflector is much more complex. In the ATR, the reflector and OSCCs must be replaced periodically because of swelling, generally at 8- to 10-year intervals.

Five ATR core internal changeouts have been completed to date. From 1970 through 1993, some of the beryllium removed from the cores of the reactors at the Reactor Technology Complex was disposed of as LLW in the SDA. Beryllium reactor waste disposed of in the SDA includes 20 beryllium blocks from ATR Cores 1, 2, and 3; nine OSCCs from ATR Cores 1 and 2; and one beryllium reflector assembly each from the MTR and the ETR. This was verified by evaluation of current storage and disposal documentation.

The ETR Critical (ETRC) was a low-power reactor that supported ETR operations. It had a beryllium reflector of nominally the same dimensions as the ETR. Beryllium reflector blocks from this reactor appear to have been removed and were probably disposed of in the SDA; although disposal records located to date do not support this conclusion. However, the ETRC beryllium reflector blocks have been included in this study to provide completeness. Because of the low power level of the ETRC, burial of its beryllium reflector in the SDA would be a neglible contribution to the C-14 inventory and to TRU inventory totals.

The majority of irradiated beryllium reflector waste—a total of 4,742 kg (10,454 lb) from the ATR, ETR, and MTR—was disposed of in the SDA in 1970, 1976, 1977, 1982, 1987, and 1993. Disposal locations and dates for the beryllium were initially obtained from shipment records. To confirm locations, vapor samples from areas indicated in shipment records were surveyed for tritium because tritium is released from the beryllium by corrosion as gas or water vapor into the surrounding soil. These confirmed locations of buried irradiated beryllium were grouted in situ as a non-time critical removal action under the National Oil and Hazardous Substances Pollution Contingency Plan (Lopez 2004). Tables listing the shipment records and identifying the locations of buried beryllium and the locations that were grouted under the removal action are in Appendix C. Two additional probable locations of beryllium disposal that were not included in the removal action have been found and confirmed by tritium surveys. These are disposals in SVR 12 that are assumed to be the ATR Core 2 reflector blocks and a small shipment of two barrels of ATR-ETR beryllium buried in Trench 57.

Previous characterizations of beryllium buried in the SDA relied on modeling alone to develop the inventories of isotopes of concern; however, certain key nuclides, such as C-14, were thought to be overestimated in the inventory and disposal of the blocks was halted. In an effort to reduce the conservatism in the inventory estimates, samples were taken from ATR beryllium blocks that were still in the ATR canal. Sampling showed that the initial models were indeed conservative with regard to C-14, but Nb-94 concentrations had been substantially underestimated because of incorrect information on Nb-93 impurity. The sampling also revealed uranium impurities in the blocks that had not been previously identified. This impurity is important because, under the neutron flux in a reactor, the uranium is transmuted to TRU isotopes. Further investigation has shown that the blocks contain a high enough TRU content to be classified as TRU waste. This classification is worth noting because it has implications for possible actions in remediating the blocks buried in the SDA.

Characterization improvements have been made for the specific isotopes C-14, Nb-94, and alpha-emitting TRU isotopes. These data were obtained through comparison of results from a new three-dimensional cross-section computer model of the ATR with measurements on samples taken from ATR reflector blocks, refinement of beginning-of-life chemical impurity estimates, and improved specificity of neutron exposure. The processes and programs used to ensure that the generated data will be suitable for the intended use are described in this report. The scope of this work also includes the development of a detailed radiological computer model for producing characterization data for the MTR and ETR reactors.

1.4 Findings

Based on the disposal dates, measurements, and calculations reported here, a total of 92.5 Ci of C-14 was disposed of in the SDA in beryllium reflector material from MTR, ETR, and ATR. Nb-94 concentrations were found to be much higher than previously expected because of higher concentrations of the Nb-93 impurity precursor. These calculations show an Nb-94 inventory of 0.0773 Ci in MTR, 0.0482 Ci in ETR, and 0.0834 Ci in the ATR beryllium at the time of disposal. Even though the C-14 inventory is lower than previously thought, the presence of higher Nb-94 concentrations indicates that the value calculated using the sum-of-fractions rule will exceed one; that is, the irradiated beryllium exceeds the regulatory requirement for Class C disposal criteria (10 CFR 61 2002). Other isotopes in the beryllium material buried at the SDA are summarized in Table 1-1 and described in Section 7.

Irradiated beryllium buried at the SDA qualifies as TRU waste. In scaling the results of calculations and measurements on ATR beryllium reflector blocks to reflectors from the ETR and MTR buried in the SDA, analysis showed that reflectors from both those reactors were TRU waste and at higher TRU isotope concentrations than the beryllium from the ATR reflectors. As indicated in Table 1-1, approximately 1.77 Ci of TRU isotopes at time of disposal were in the reflectors from ETR and MTR. Some increase in activity levels is a consequence of lower neutron flux in ETR and MTR beryllium that burned away fewer TRU isotopes produced while in the core. Also, the longer time—in comparison with ATR beryllium—that ETR and MTR reflectors have been out of their reactors has allowed greater build-up of TRU isotopes from the precursors.

Improved computer models and cross sections show good agreement between predictions and measurements, thus these tools developed for characterizing ATR beryllium should give good predictions for beryllium from ETR and MTR. These models and results are discussed in Section 6.

Transuranic waste isotope concentrations in irradiated beryllium change with time. Decay of non-TRU precursors causes TRU activity generally to increase for decades after the beryllium has been removed from the reactor. Burnout of some isotopes means that the greatest TRU activity will not necessarily be where neutron fluence is highest.

Further research into shipment documents and tritium surveys has resulted in changes to beryllium disposal dates and locations as reported in previous versions of this report. In particular, MTR beryllium is now believed to have been disposed of in two separate campaigns in 1970 and 1977 and beryllium from ATR Core 2 is now believed to have been disposed of in SVR 12 in 1982. These changes will affect the calculated radionuclide inventory at the time of disposal but do not impact the calculated radionclide inventory at the end of irradiation.

Table 1-1. Summary of the Advanced Test Reactor, Engineering Test Reactor, and Materials Test Reactor irradiated beryllium reflector waste disposed of in the Subsurface Disposal Area.

Reactor and Beryllium Waste Buried	Initial Irradiated Date	Final Irradiated Date	Core	Be Metal Mass (g)	Metal Volume (m^3)	Disposal Date	Disposal Location	Total C-14 (Ci)	C-14 Concentration (Ci/m^3)	Transuranic Concentration (nCi/g)	Total Transuranic (Ci)
Materials Test Reactor[a]	3/31/52	7/3/69	1	~616,000	~0.333	1970	Trench 52	9.01	27.1	694	0.426
			1	~1,384,000[b]	~0.747	1977	Trench 58	20.2	27.0	854	1.18
Engineering Test Reactor	10/15/57	3/1/70	1	~624,000[c]	~0.337	1970	Trench 54	21.7	64.4	257	0.160
Advanced Test Reactor											
Eight reflector blocks	2/1/68	9/9/72	1	651,360	0.352	1976	Trench 58	7.81	22.2	319	0.210
Six reflector blocks	2/5/73	4/11/77	2	488,520	0.264	1982	Soil Vault Row 12	5.83	22.0	327	0.160
Six reflector blocks	8/9/77	2/2/86	3	488,520	0.264	1993	Soil Vault Row 20	12.0	45.3	379	0.185
Nine outer shim control cylinders	2/5/77	4/11/77	1 and 2	489,881	0.2648	1987	Soil Vault Row 17	15.9	60.1	297	0.145
Total				4,742,281	2.562			92.5			2.47

a. The breakdown of MTR beryllium was based on reported shipment weights in 1970 and 1977.
b. 177,887 MWd is reported by the *MTR Progress Report Cycle No. 295* (Ford et al. 1969). An alternate value of 179,329 MWd (71,322 + 108,000) can be determined from data shown on pages 10 and 11 of *Characterization of the Materials Testing Reactor* (Rolfe and Wills 1984). Rolfe and Wills (1984) also report that the total operating history of MTR (to August 21, 1970) was about 180,000 MWd. The MTR beryllium mass is based on dimensional data (IAEA 1959) and a beryllium density of 1.85 g/cm^3. The uncertainty in the MTR beryllium mass is approximately ±14%.
c. The final irradiation date for ETR before removal of the original beryllium reflector is estimated to be March 1, 1970. As noted on page 35 of *Characterization of the Engineering Test Reactor Facility—September 1982* (Kaiser et al. 1982), the original beryllium reflector was replaced sometime in March 1970 (but the exact day of the month is not reported). The total number of MWd of ETR operation to March 1970 has not been identified. However, the number of MWd of ETR operation to January 11, 1970, is reported as 374,498 MWd on page 8 of *ETR Operations Branch Progress Report for Cycle No. 105 November 7, 1969—January 11, 1970* (Smith et al. 1970). Also, Kaiser et al. (1982) report 487,728 MWd of ETR operations through 1972. The estimated value of 380,000 MWd is based on an extrapolation of the January 11, 1970, value. The uncertainty associated with this value is ±2%. The ETR beryllium mass is calculated from dimensional data (IAEA 1964; Tobias 1969) and assumes a beryllium density of 1.85 g/cm^3. The uncertainty in the ETR beryllium (reflector) mass is estimated to be about ±1%.

ETR = Engineering Test Reactor
MTR = Materials Test Reactor
MWd = megawatt-day

1.5 Overview of Methodology and Assumptions

The characterization methodology calculates radionuclide concentrations in irradiated beryllium reflector materials (blocks and OSCCs) from ATR based on new characterization data and improved computer modeling of beryllium reflectors stored in the ATR canal. Computer modeling focused on beryllium Block 010R from ATR Core 3 that is currently stored in the canal. The characterizations and validation results of these beryllium reflector materials are then used to scale characterization data for the beryllium components from MTR and ETR.

The logical flow of characterization activities is presented in Figure 1-3. It identifies separate tasks and captures the complex process logic that has evolved over the course of characterization efforts specific to irradiated beryllium generated by ATR, MTR, and ETR. Characterization efforts are focused on beryllium from ATR rather than beryllium from MTR and ETR because of the following:

- More complete operational records are available
- Stored blocks are available for sampling
- A disposal path must be identified for stored ATR beryllium blocks.

ATR beryllium characterization centers on two codes used for model calculations: Monte Carlo N-Particle Version 4B (MCNP4B) and Oak Ridge Isotope GENeration and Depletion Code Version 2 (ORIGEN2). These models have been validated against measured assay data to ensure adequacy, accuracy, and confidence. These models, and the radionuclide inventories produced by these models, are the bases for the activation product concentrations for C-14, TRU isotopes, and other reported estimates of radioisotopes in the irradiated beryllium buried in the SDA.

To characterize MTR, ETR, and ATR beryllium, ATR beryllium Block 010R is assumed to be similar—both physically and radiologically—to the beryllium materials in all three reactor cores. This assumption is logical for the ATR beryllium because the beryllium blocks remaining in the canal are from ATR Cores 2, 3, 4, and 5, and the blocks buried in the SDA are from ATR Cores 1, 2, and 3. However, for MTR and ETR beryllium, a similar assumption is not warranted as strongly because the beryllium supplier is not known and measured data do not exist for MTR and ETR beryllium. The operational time period of MTR and ETR partially overlapped that of procurement for the ATR beryllium; therefore, the assumption is that beryllium procured during concurrent time periods would have been supplied by the same manufacturer and supplier and would have similar characteristics. Sections 3 and 4 provide a more in-depth development of the relationships among beryllium reflector materials in MTR, ETR, and ATR.

1.6 Report Organization

The remaining sections of the report are summarized below:

- Sections 2, 3, and 4 summarize the configuration and operational histories of ATR, MTR, and ETR, respectively.

- Section 5 discusses future generation and possible future disposals of beryllium reflector waste from ATR, ETR, and MTR as remote-handled LLW in the SDA active pit.

- Section 6 describes calculation techniques and tools used to predict activation product concentrations and radionuclide inventories for ATR beryllium Block 010R.

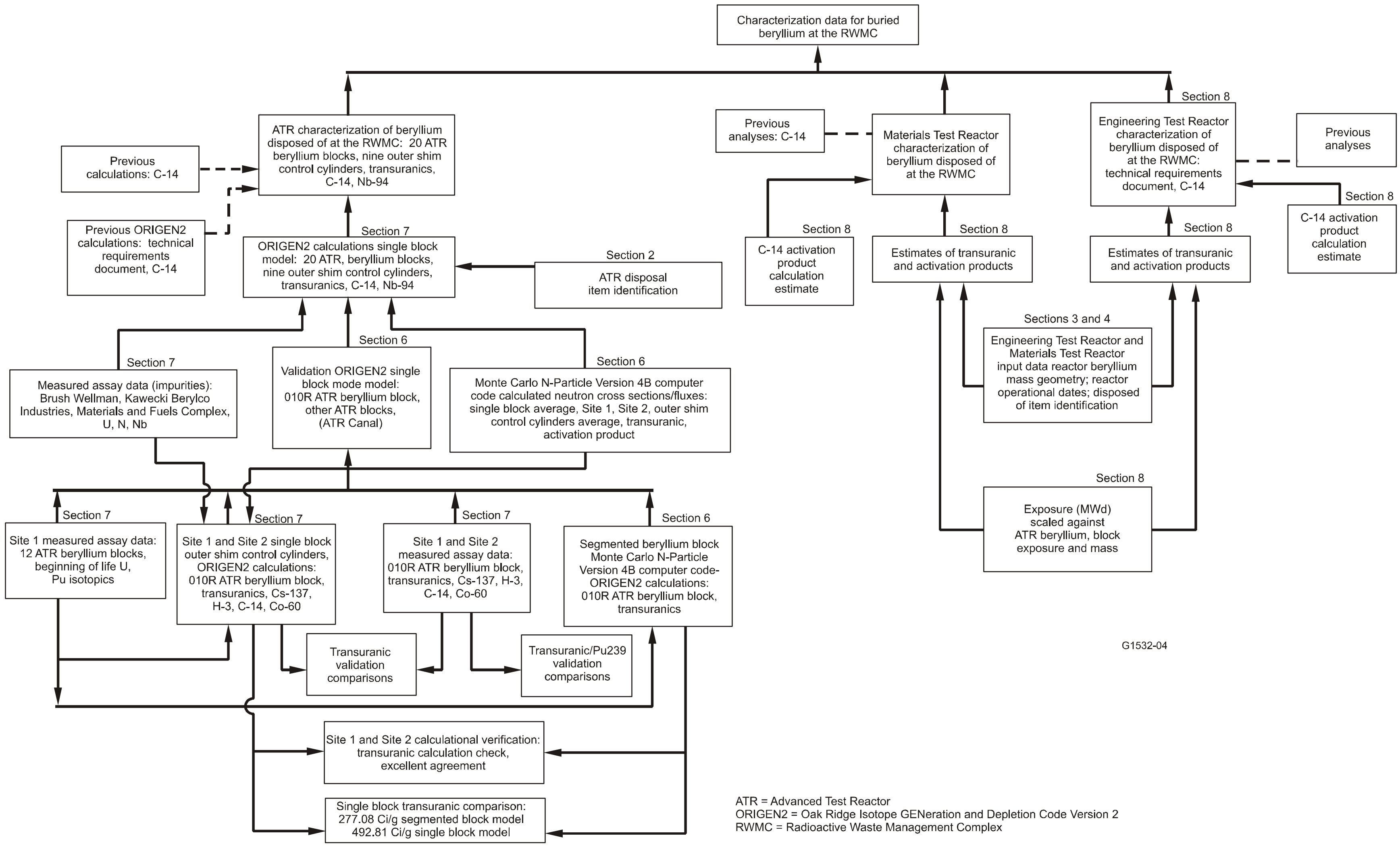

Figure 1-3. Characterization methodology for beryllium components from the Advanced Test Reactor, Materials Test Reactor, and the Engineering Test Reactor that are buried in the Subsurface Disposal Area.

- Section 7 describes the ORIGEN2 input models that calculated radionuclide concentrations in the beryllium reflector blocks disposed of in the SDA. Calculated radionuclide inventories for the beryllium are also in this section.

- Section 8 presents conclusions and recommendations for characterizing beryllium reflector material from MTR, ETR, and ATR buried in the SDA.

- Section 9 contains the references cited throughout the report.

- Appendix A contains assay test data on beryllium from the ATR purchased from Kawecki Berylco Industries (KBI).

- Appendix B contains analytical test data from the Materials and Fuels Complex (formerly Argonne National Laboratory-West) on the advanced test reactor beryllium samples).

- Appendix C contains beryllium shipment records and disposal locations.

- Appendix D contains a comparison of grouted and ungrouted beryllium.

2. BERYLLIUM MATERIAL FROM THE ADVANCED TEST REACTOR

This section describes the beryllium reflector material used in the ATR reactor and its physical configuration, weight, neutron exposure, and the location in the reactor. This section also reviews waste disposal documents that supported identifying and numbering the items of beryllium reflector material from ATR disposed of in the SDA. Results from this analysis of ATR beryllium are later used to estimate inventories in other beryllium buried in the SDA.

2.1 Description and Operational History

The ATR reactor has a beryllium reflector, as do some other research reactors, to intensify the neutron flux in the core. The ATR reflector consists of a set of eight blocks and 16 OSCCs. A photograph of a reflector block is shown in Figure 2-1. Each block has a mass of about 81,420 g (179.5 lb) after machining. Hence, a set of eight blocks would contain 651,360 g (1,436 lb) of beryllium. Block metal volume after machining is 0.044 m^3. Each block is 129.5 cm (51 in.) long (Erickson 1966). The cross-section of a block can be fit in a circle nominally 51 cm (20 in.) in diameter. Relative placement of these blocks and OSCCs in the ATR core is shown in Figure 2-2.

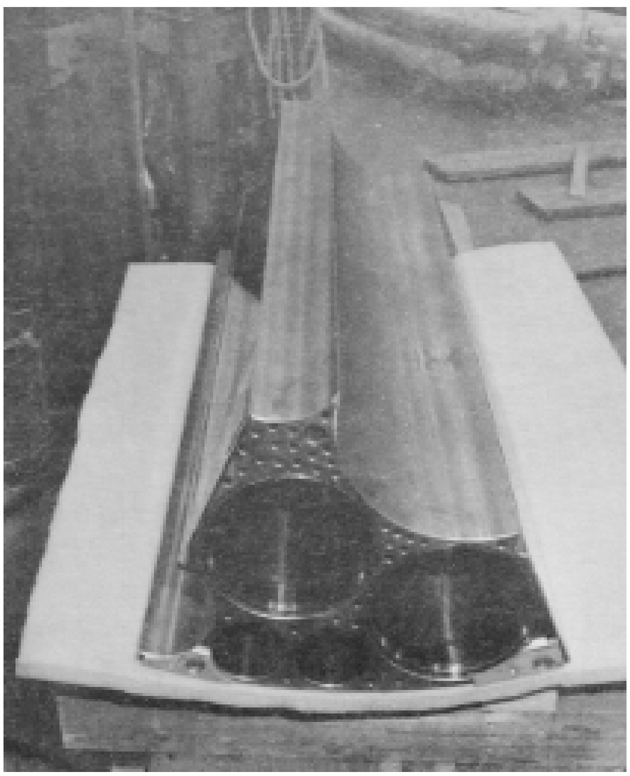

Figure 2-1. Photograph of a beryllium reflector block from the Advanced Test Reactor.

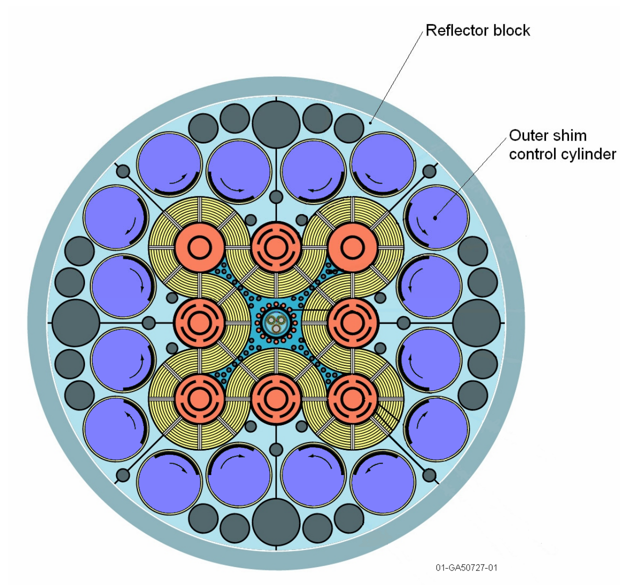

Figure 2-2. Cross-sectional view of placement of beryllium blocks and outer shim control cylinders in the core of the Advanced Test Reactor.

The use of beryllium in reactors generates substantial amounts of He-4, He-3, and H-3 that accumulate and cause the beryllium to swell, thus requiring replacement of the beryllium blocks periodically. Since the ATR began operation in 1967, the beryllium reflector blocks have been replaced in five internal changeouts of the core in 1972, 1977, 1986, 1994, and 2004. The OSCCs, installed during original plant construction, were reused for reflector Core 2, and changed out in 1977 (Logan 1999). Including the original reflector, five beryllium reflectors (40 blocks) have been removed from the ATR. The sixth beryllium reflector currently remains in the ATR.

OSCCs are circular cylinders 18.4 cm (7.25 in.) in diameter and 119 cm (46.8 in.) long. They hold hafnium plates that control reactor flux. OSCC weight in air, excluding the weight of attached hafnium components, is 57 kg (125 lb). Early OSCCs were single pieces, but later they were built from three shorter cylindrical sections. Each core has 16 OSCCs (48 segments). Sixteen OSCCs from Core 1 were reused in Core 2. Placement of the OSCCs in the ATR core is shown in Figure 2-2.

2.2 Chemical Impurities in Reflector Material

Beryllium reflector material used in the ATR beryllium blocks has come primarily from two suppliers:

- KBI provided the reflector blocks for Cores 1 through 4. Defunct since 1979, KBI was located in Reading, Pennsylvania.

- Brush Wellman produced the blocks in Core 5 and Core 6 (currently in the reactor).

The elemental composition of beryllium varies from manufacturer to manufacturer depending on the source of the beryllium ore, the amount of blending of different ore sources, the amount of recycled beryllium used from unknown sources, and to some degree, the manufacturing process used to grind beryllium particles to the correct size and press the beryllium billets. Beryllium reflector material used in ATR OSCCs also has come from both of these suppliers. However, all of the nine OSCCs disposed of in the SDA are known to be provided by KBI (because they were from Cores 1 and 2 and were provided in the initial procurement). Specific ATR reflector material and chemical impurities used for radiological characterization modeling is fully described in Section 7.

2.3 Disposal of Beryllium Material from the Advanced Test Reactor in the Subsurface Disposal Area

To validate disposal of ATR beryllium material in the SDA, the following five sources were reviewed: Logan (1999), Moncur (1998), the Radioactive Waste Management Information System (RWMIS) waste disposal database, individual waste shipment records (Idaho Operations Office Waste Disposal Request and Authorization Record), and the *Sampling and Analysis Plan for ATR Beryllium* (Haney 1999). None of these individual references offers a comprehensive or correlated inventory of ATR beryllium items disposed of in the SDA. Correlation of disposal information between each of the references to develop a comprehensive inventory remains incomplete because neither the RWMIS database records nor the individual disposal and shipment records have enough information to clarify disposal of specific ATR beryllium materials. RWMIS and shipment records are either incomplete or inconsistent in identifying specific beryllium materials disposed of and dates of disposal.

Gay (Logan 1999) used a mass balance approach to determine the beryllium blocks and OSCCs that were buried in the SDA. This approach inventoried the beryllium reflector components remaining in the ATR canal and subtracted this amount from the total activated material that was generated over the operating period of the ATR. This approach correctly identified the total number of 20 beryllium blocks and nine OSCCs by core number that were disposed of in the SDA. However, none of the references gave serial numbers from individual beryllium reflectors, except for the six blocks from Core 3.

In 1999, samples were collected from the 12 beryllium blocks in the ATR canal to analyze for C-14 and nitrogen. This sample collection is documented by Haney (1999) and associated laboratory logbook entries. Individual serial numbers of the beryllium blocks were recorded, allowing development of a core-specific inventory of reactor lobe positions of beryllium blocks to be used in calculating the isotopic concentration characterization both for each of 12 blocks disposed of in the ATR canal in 1999 and for 20 blocks disposed of in the SDA. The new inventory of block serial numbers revealed that previous information reported by Gay (Logan 1999) and used in subsequent references was in error. Previous reports indicated that eight blocks from Cores 1 and 2 and four blocks from Core 3 had been disposed of. However, distinguishing characteristics of the beryllium block cores (i.e., saw cuts) observed during sample collection and reported in the laboratory logbook indicated that two of the blocks remaining in the canal only could have been from Core 2. The new information indicates that six blocks (not eight blocks) from Core 2 were disposed of in the SDA.

Some questions remain concerning when and which specific Core 2 blocks were disposed of in the SDA because the references do not identify the blocks by serial number. The sampling and analysis plan for ATR beryllium (Haney 1999) indicates that two blocks from Core 2 remain in the ATR canal. This confirms that six blocks from Core 2 were disposed of in the SDA. A shipment record documents the disposal of 600 lb of irradiated beryllium from ATR in October 1982. Although this is not sufficient to account for all Core 2 blocks, the assumption is that all six Core 2 blocks were shipped to the SDA at this time and disposed of in approximately the same location. This assumption is consistent with the shipping activities for Core 1 blocks in 1976 and Core 3 blocks in 1993.

The OSCC beryllium has not been sampled or analyzed. SDA disposal dates, mass balance approach, and canal operator information indicate that the nine OSCCs disposed of in the SDA could have come only from Cores 1 and 2 (Core 1 OSCCs were reused in Core 2). No known records exist and no serial numbers are recorded for those nine OSCCs.

This analysis indicates that the ATR disposals of beryllium material in the SDA were as follows:

- November through December 1976: eight blocks (from Core 1) and no OSCCs.
- October 1982: six blocks (from Core 2) and no OSCCs.
- August through September 1987: no blocks and nine OSCCs (from Cores 1 and 2; note that Cores 1 and 2 used the same OSCCs).
- May through June 1993: six blocks (from Core 3) and no OSCCs.

Therefore, a total of 20 blocks and nine OSCCs were disposed of in the SDA from 1976 through 1993.

3. BERYLLIUM MATERIAL FROM THE MATERIALS TEST REACTOR

This section presents information on the beryllium reflector materials used in the MTR. The description includes the physical configuration, weight, neutron exposure, and the location in the reactor. The manufacturer of the reflector and the relationship to the ATR beryllium reflector is evaluated. Waste disposal documents support the assumption that the first MTR beryllium reflector material was most likely disposed of in the SDA.

3.1 Description and Operational History

A cross section of the MTR reactor is depicted in Figure 3-1. The shaded segments represent the beryllium reflector. It comprised a large number of pieces, each 99 cm (39-3/8 in.) high. Some were in the general shape of fuel elements and were located with the fuel elements inside the core. Others were shaped prismatic blocks that fit within the spaces between the various MTR structures. Some were essentially rectangular, others were wedges, and many had holes for test capsules. They also had a large number of flow passages for the water coolant. The estimated weight of the beryllium reflector is about 2,000 kg (4,410 lb).

The first beryllium reflector functioned in the reactor from when it became operational on March 31, 1952, until July 3, 1969, when the reactor was shut down to change the fuel configuration (Rolfe and Wills 1984).

Beryllium can be extracted from many different minerals, mined in a number of different countries, notably Kazakhstan, Brazil, Zambia, Argentina, Australia, Angola, and Rwanda (Floyd and Lowe 1979). In recent decades, nearly all U.S. beryllium has been manufactured from Spor Mountain bertrandite ore mined at the Brush Wellman mine in Delta, Utah. The ore from Spor Mountain is known to contain relatively high levels of uranium and gold impurities. Production from the Delta, Utah mine did not start until 1969, 12 years after the ETR operations began and after the MTR operations had essentially finished. Therefore, the ETR and MTR beryllium could not have been produced from the Brush Wellman mine at Spor Mountain.

Beryllium with exceptionally low uranium impurity is known to come from the Ulba Metallurgical Plant in Kazakhstan, but the United States would not have been purchasing this strategic material from the Soviet Union in the early 1950s. Likewise, the United States probably would not have been buying it from China. More likely, either Brush Wellman or KBI would have furnished the ETR beryllium rather than the French or Indian companies. Therefore, the most likely source of the MTR and ETR beryllium metal was KBI.

Before the discovery of the Spor Mountain bertrandite deposit in 1959, most of the domestic beryllium reserves were as beryl and other minerals in spodumene deposits in South Carolina (Floyd and Lowe 1979). The MTR and ETR beryllium could have come from South Carolina, though most of the world-market beryllium in those years came from Brazil (Hanafee 1999). The ETR and MTR beryllium probably would have similar uranium impurities regardless of whether it was furnished by Brush or by KBI. A further point is the propensity of a consumer to stay with the same source of supply for many critical components. Hence, KBI or one of its predecessors is assumed to have furnished the MTR beryllium.

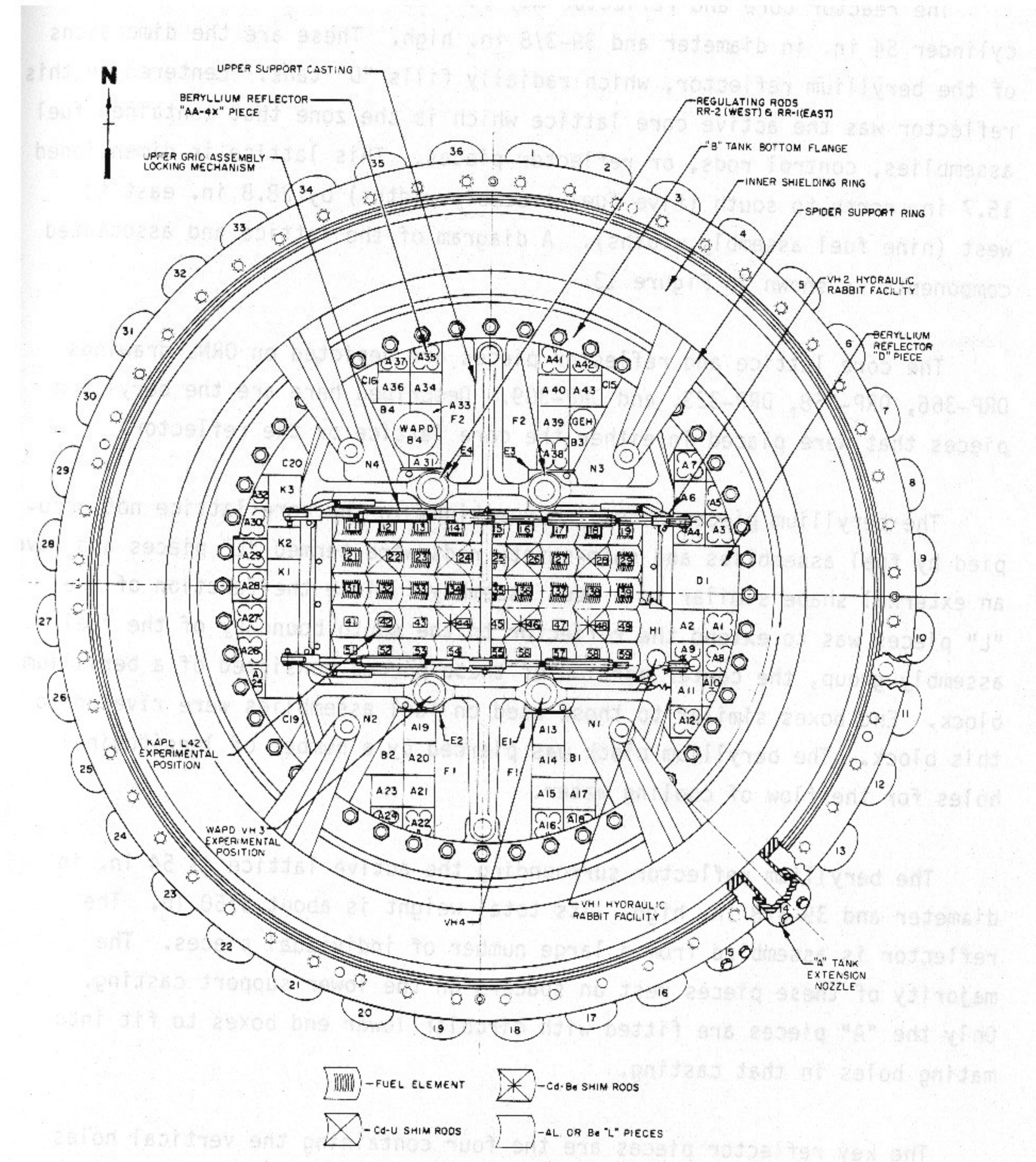

Figure 3-1. Configuration of the beryllium reflector for the Materials Test Reactor.

Measurements of selected impurities were made on beryllium samples from KBI-furnished ATR reflector blocks (Appendix A). The blocks sampled came from Cores 2, 3, and 4, spanning the years from the late 1960s to 1994. Those samples showed a uranium concentration of 28.5 ± 5.8 weight parts per million (wppm). Only one sample included uranium levels in the 40- to 100-wppm range typical of the Spor Mountain ores (41 wppm from Core 2). Combining the sample-to-sample variation, the uncertainty in the measurements of 10%, and the uncertainty of the source of the beryllium metal for the MTR, a typical beginning-of-life uranium concentration of 30 ± 10 wppm would be present. For issues relating to

TRU waste, uranium is the only impurity of concern, though others, such as N-14 and Nb-93, certainly will affect the β^- emitters for determination with respect to the remedial investigation risk assessment for the OU 7-13/14 RI/FS. Concentrations in those samples were 205 ± 41.2 wppm for N-14 while concentrations were 11.7 ± 8.3 wppm for Nb-93.

3.2 Disposal of Beryllium Waste from the Materials Test Reactor

Rolfe and Wills' report (1984) was reviewed as a reference evaluating the disposal history of beryllium from MTR. Then RWMIS and Idaho Operations Office Waste Disposal Request and Authorization Records (disposal records) were reviewed to validate the disposal information in the references and to ascertain if more detailed information might have been listed on the actual disposal records. Best available information indicates that the four beryllium disposals from MTR were made from April through May 1970 and nine shipments were made from May through June 1977. These shipments contained one entire reflector from Core 1. Shipment weights indicate approximately 31% of MTR beryllium was disposed of in 1970 and the remainder was disposed of in 1977. The shipments corresponding to the disposal of beryllium from MTR are listed in Appendix C.

4. BERYLLIUM MATERIAL FROM THE ENGINEERING TEST REACTOR

This section describes the beryllium reflector material used in the ETR reactor and compares beryllium from ETR with beryllium from ATR. A review of waste disposal documents supports the assumption that the first ETR beryllium reflector material was most likely disposed of in the SDA.

4.1 Description and Operational History

The configuration and arrangement of the beryllium reflector from ETR is shown schematically in Figure 4-1. The original ETR reflector consisted of four blocks, each 11.4 × 88.9 × 95.3 cm (4.5 × 35 × 37.5 in.), that were linked together at the corners to completely surround the core laterally. The link plates were probably made of beryllium and had lifting balls on the ends to facilitate removal. Each of these blocks was penetrated by either seven or nine vertical test holes (seven on the west side, nine on the other three sides). Each block had two 4-cm (1.6-in.) diameter holes, and the rest were 3.3 cm (1.3-in.) in diameter. Each block also had a large number (98 or 99) of 6.3-mm (1/4-in.) coolant passage holes.

The first ETR reflector was removed in March 1970 and disposed of in the SDA. The second reflector was essentially the same configuration, except it was subdivided into ten vertical layers (40 pieces in all). The ETR was placed in standby status in 1980 and never restarted. The second beryllium reflector remains in the ETR today.

The mass of the beryllium from ETR Core 1 is estimated to be 624 kg (1,376 lb) from the geometry and beryllium specific gravity of 1.85. That does not include filler pieces in the test holes or the lifting balls on the link plates. Thus, each block in the first reflector would weigh about 156 kg (344 lb).

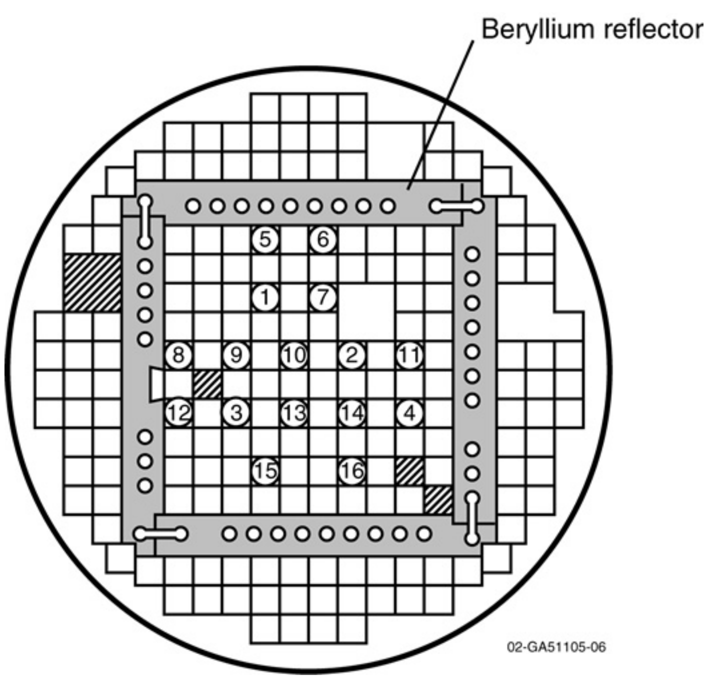

Figure 4-1. Schematic of beryllium reflector in the Engineering Test Reactor.

4.2 Chemical Impurities in Reflector Material

The initial composition of impurities in the beryllium in ETR is not known. Typical impurity levels are strongly correlated with the source of the ore from which the beryllium is reduced and less so with the manufacturing method. Based on a comparison, the beryllium for the ETR probably was manufactured from the same beryl ore used by KBI for the early ATR reflector blocks.

The discussion provided for the MTR beryllium in Section 3 also is applicable to ETR beryllium.

4.3 Disposal of Beryllium Waste from the Engineering Test Reactor

The shipping records in INL's RWMIS database and the Idaho Operations Office Waste Disposal Request and Authorization Records (i.e., disposal records) that pertain to the disposal of beryllium waste were reviewed. All of the reviewed information indicates that most, if not all, of the beryllium waste from the first ETR core was disposed of in the SDA. The only shipment from ETR whose record specifically mentions beryllium waste was disposed of in Trench 54 during December 1970. Additional high curie shipments from ETR containing core components were also disposed of in Trench 54 in this time period. Therefore, Trench 54 is assumed to contain the remaining beryllium waste. Shipments assumed to contain beryllium waste from ETR are listed in tables in Appendix C. The second ETR reflector remains in the ETR reactor core.

4.4 Engineering Test Reactor Critical Facility

The ETRC facility was located in TRA-654 in the southeast corner of the MTR Reactor Services Support Building (TRA-635), next to the ETR building. The ETRC was a full-scale, low-power mockup of the ETR and had a beryllium reflector that was a replica of the one in ETR.

Kaiser et al. (1982) state "...the [ETRC] canal was drained and left covered with plywood sheeting…" and that "all radioactive components have been removed from the facility." These statements indicate that the beryllium reflector had been removed. Based on this statement, the ETRC beryllium reflector is assumed to be disposed of in the SDA. However, no reference to disposing of an ETRC beryllium reflector was found in the INL's RWMIS, or other disposal records or references investigated. Since Kaiser et al. published in 1982, the ETRC beryllium reflector is assumed to have been disposed of before that date.

The ETRC first became operational on May 20, 1957 (Burdick and Henscheid 1958). While it was capable of 75 kW_{th}, the high-power trip point was never set above 50 kW_{th} (INEL 1979). The average neutron flux in the fuel elements was 2.72×10^8 n/cm^2/second, and the average power was only 216 W_{th} (Burdick and Parry 1958). The values contrast markedly with the high flux value in the ETR, which was 8×10^{14} n/cm^2/second at the rated power of 175 MW_{th} (Kaiser et al. 1982). Thus, the average ETRC flux was more than two million times lower than that for ETR.

Approximately two-thirds of the beryllium used for the ETRC reflector came from modified beryllium pieces that were available from another facility. The remaining one-third of the reflector was made from sintered beryllium oxide and was contained in aluminum cans.

The entire irradiated beryllium reflector of MTR was assumed to be disposed of in the SDA; also assumed was that none of the beryllium had been reused in the ETRC. In addition, the ETRC beryllium reflector was assumed to have had no preirradiation history or preradiological characteristics. The only neutron activation in the ETRC beryllium reflector was from the ETRC itself and not from prior MTR activation.

While the detailed operating history of ETRC is not known, a highly conservative assumption is that it operated only for the same period as the ETR. The ETRC duty factor was probably only 1%. The ETR first became operational in October 1957, with full power being achieved in 1958, and continued until 1981. The ETR reflector was replaced in March 1970.

Hence, the neutron fluence in the ETRC reflector was at most one millionth of that in the ETR reflector and probably only one hundred-millionth (10^{-8}). A simple activation model for typical beryllium from that era in an average neutron flux of 2.72×10^6 n/cm^2/second gives TRU activity levels essentially just those for the inherent uranium impurity plus an equivalent level of Pa-234 that saturates after about 7 months. Increasing the neutron flux by a factor of 100, corresponding to operation at a power in excess of 20 kW$_{th}$, does little to change that result, only extending the time required to reach steady state. Based on this analysis, no issues relating to TRU waste are associated with the ETRC reflector, the total activity being only about 0.02 nCi/g.

While not explicitly discussed here, activation of the beta emitters C-14 and Nb-94 also would be well below any threshold value of concern. They were only twice the greater-than-Class-C thresholds (10 CFR 61 2002) at the full ETR power levels, and their production corresponds very closely to total neutron fluence.

5. FUTURE BERYLLIUM WASTE REQUIRING DISPOSAL

The assumption for test program planning for the ATR—an operating test reactor—is that the reactor will continue to operate at past power levels through the year 2050. This section discusses the beryllium to be disposed of in the future.

5.1 Advanced Test Reactor: Future Generation and Disposal of Beryllium Waste

In the past, beryllium reflector blocks have been changed out at intervals of 8 to 10 years. In the five core internal changeouts completed to date, 40 blocks and 64 OSCCs have been generated. Of these, 20 blocks and nine OSCCs have been disposed of in the SDA (note that Core 2 reused the OSCCs from Core 1). The fifth core internal changeout (replacing Core 5 with Core 6) was completed in early February 2005. With an average interval of nine years for changeouts, approximately five more cores will be changed out through 2050. These additional five changeouts will generate 40 more beryllium blocks and 80 OSCCs. Twenty blocks and 55 OSCCs generated from previous core internal changeouts are currently in the ATR storage canal. Therefore, total beryllium waste requiring disposal in the future is estimated at 60 blocks and 135 OSCCs. In addition, beryllium plugs—used from core to core—that fill the test holes in the reflector blocks for experiments will require disposal at facility closure. The number of hole fillers and their mass is not known at this time.

New characterization information for the beryllium blocks and OSCCs has changed the waste stream classification of the ATR beryllium waste from LLW to TRU waste. If the uranium chemical impurity in the future-procured beryllium reflectors remains similar to beryllium used in the past, all future-irradiated ATR beryllium will be classified as TRU waste. This waste stream designation will prohibit future beryllium disposal in the SDA as LLW.

In addition, beryllium from the Advanced Test Reactor Critical (ATRC) facility—a low-power replica of the ATR core—must be characterized and eventually disposed of in the future. This facility is now operational and determines in advance the nuclear characteristics of experiments programmed for irradiation in the ATR. The ATRC beryllium reflector is a full-scale replica of the ATR core and includes OSCCs and hole fillers. However, because of the very low power history for the ATRC, the beryllium waste from this reactor possibly will be suitable for LLW disposal at decommissioning. The ATRC is assumed to be operational for the same period as the ATR (i.e., until 2050). The original ATRC beryllium reflector is still in the reactor and is expected to remain so during ATR's operational period. Sufficient flux exposure of the beryllium to cause the reflector blocks to be classified as TRU waste is highly unlikely because of the low power levels and infrequent periods at which ATRC operates.

An option addressed in "Radioactive Waste Management" (DOE O 435.1 2001) allows special decisions for disposal—given technical justification—based on performance assessments. Final disposal approval is required from the U.S. Secretary of Energy with concurrence from the administrator of the U.S. Environmental Protection Agency. The decision process evaluates whether the degree of isolation required for TRU waste is necessary for a specific waste. If that degree of isolation is not required, then the waste can be exempted from the TRU waste classification and reclassified as LLW. Though beryllium waste may not require the degree of isolation generally required for TRU waste, pursuing an exemption will be difficult because no precedent for seeking such an exemption or experience with the process itself has been reported. Therefore, receiving such an exemption would be in doubt for disposal of ATR beryllium.

Using the LLW disposal facility at the SDA to dispose of beryllium reflector materials is not anticipated because the current and future ATR beryllium reflector waste is classified as TRU waste, and it does not meet the disposal criteria for LLW in the SDA. In addition, the SDA LLW disposal facility will be volume-filled by 2020 or administratively closed in 2009, a full 30 years before the anticipated decommissioning of ATR and ATRC.

5.2 Materials Test Reactor: Future Generation and Disposal of Beryllium Waste

The MTR has been deactivated; however, the second beryllium reflector is still in place. Beryllium waste from MTR will be a concern for future deactivation, decontamination, and decommissioning, which is scheduled to be finished by 2012. In an MTR characterization report, Rolfe and Wills (1984) indicate that a beryllium core changeout occurred in 1969, near the end of the operational life of the facility; therefore, the beryllium core now in place has not been exposed to large amounts of neutron flux. Rolfe and Wills (1984) did address characterization of the beryllium reflectors, but not sufficiently to allow TRU waste determination. Evaluating the remaining MTR beryllium reflector is beyond the scope of this report; however, because of low neutron exposure, MTR beryllium may not be TRU waste. Further characterization will be required to develop a final waste stream determination at the time of decommissioning. Disposal of the MTR beryllium reflector as LLW in the SDA LLW disposal facility is not expected to be an option because the SDA LLW disposal facility will be volume-filled by 2020 or administratively closed in 2009.

5.3 Engineering Test Reactor: Future Generation and Disposal of Beryllium Waste

The ETR reactor has been deactivated. Beryllium waste generation in the ETR will be a concern for future deactivation, decontamination, and decommissioning, which is scheduled to begin after 2006. In an ETR characterization report, Kaiser et al. (1982) indicate that the initial beryllium core was changed out in 1970 and the new beryllium core remained in the operating reactor through 1980 when ETR was shut down. During this operational period, the new beryllium reflector experienced significantly low neutron fluence, which may not have been sufficient to cause the reflector to become TRU waste. Kaiser et al. (1982) did address characterization of the beryllium reflector, but not sufficiently to allow a TRU waste determination to be made. Evaluating the remaining ETR beryllium reflector characterization is beyond the scope of this report; however, because of the low neutron exposure, it is possible the beryllium reflector may be LLW. Further characterization will be required to develop a final waste stream determination at the time of ETR decommissioning. Disposal of the ETR beryllium reflector in the SDA LLW disposal facility is not expected because this disposal facility will be volume-filled by 2020 or administratively closed in 2009. The ETR could undergo deactivation, decontamination, and decommissioning in 2012, which is before the SDA late-closing date of 2020, but after the administrative closure date of 2009. The assumption for this report is that the ETR beryllium will not be disposed of in the SDA. Though the beryllium reflector may remain classified as remote-handled LLW in the future, it will probably not be disposed of at the INL Site because these disposal facilities are scheduled to close within 5 years according to the 2012 Plan (DOE-ID 2002b). The ETRC, a low-power companion reactor to ETR, supported ETR operation by determining in advance the nuclear characteristics of experiments programmed for irradiation in ETR. The building that housed the reactor has had all the radioactive materials removed. The beryllium reflector is assumed to be among these radioactive materials and to have been disposed of previously in the SDA. No future beryllium reflector waste will be generated by ETRC.

5.4 Advanced Test Reactor: Future Procurement of Beryllium

A possibility for future procurement of beryllium that may benefit future beryllium disposal is revising the ATR beryllium procurement specification. Revising the procurement specification may be technically and economically feasible to reduce chemical impurity levels in future beryllium material for those elements that complicate waste disposal. Procuring beryllium for ATR reflectors has a long lead time and the next procurement process will start some time near 2007.

Because ARTC is a low-power nuclear reactor, the existing beryllium reflector is assumed not to require replacement during the operational time period (i.e., until 2050) of the ATR reactor.